Monte Carlo simulation of circular grain growth

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Monte Carlo simulations have been carried out to study the effect of temperature on the growth kinetics of a circular grain. This work demonstrates the importance of roughening fluctuations on the growth dynamics. Since the effect of thermal fluctuations is stronger in d=2 than in *d*=3, as predicted by *d=3* theories of domain kinetics, the circular domain shrinks linearly with time as $A(t)=A(0)-\alpha t$, where $A(0)$ and A(t) are the initial and instantaneous areas, respectively. However, in contrast to d=3, the slope α is strongly temperature dependent for $T \geq 0.6T_C$. An analytical theory which considers the thermal fluctuations agrees with the T dependence of the Monte Carlo data in this regime, and this model show that these fluctuations are responsible for the strong temperature dependence of the growth rate for d =2. Our results are particularly relevant to the problem of domain growth in surface science.

1 Introduction

Since the microstructure of materials plays an important role in physical and mechanical properties of engineering materials, one of the major objectives of materials science is controlling of the grain structure by grain deformation, recovery, recrystallization, and grain growth in materials [1]. A theoretical basis for understanding the grain growth is laid down in the classic papers of Smith (1948, 1952) and Burke and Turnbull (1952) [2]. The Burke and Turnbull analysis assumes that the mean behaviour of the whole array of grains can be inferred from the migration rate of boundary part and does not consider the interaction between grains. Also Moldovan et al. [3] have revealed a coupling phenomenon between the mechanism of grain boundary migration and the grain rotation coalescence during grain growth. Another subject of grain growth developed mainly by Hillert [4] was the uniformity of the microstructure during the growth phenomena consisting of normal grain growth and abnormal grain growth. In the normal growth, the microstructure exhibits a uniform increase in grain size whereas in abnormal grain growth the size of a few grains increases rapidly so that the normalized grain size distribution is no longer time invariant [5,6].

In other works experimental interest has focused on the lattice structures observed for atoms that are adsorbed on solid surfaces. At high temperatures, the adsorbed atoms are in a disordered (gas or liquid) phase on the surface while at low temperatures, they form periodic structures. If the surface density of adsorbed atoms is less than the surface density of substrate atoms, the periodic structures formed have lattice constants that are larger than the substrate lattice constants and are known as superlattices. For physisorbed systems, such as rare-gas atoms adsorbed on graphite, superlattices that are either commensurate (i.e., the superlattice periodicity is an integral multiple of the substrate lattice constant) or incommensurate with the substrate periodicity have been observed. On the other hand, the stronger interactions that exist in chemisorbed systems usually result in a commensurate overlayer of atoms [6]. In

addition to surface studies, similar superlattice structures have also been observed in intercalation compounds, where the large spacings between intercalant layers can result in quasi-two-dimensional behaviour.

In this paper we present a theoretical study of the time and temperature dependence of domain sizes for ordering systems with several degenerate equilibrium states. The system is quenched from a high-temperature disordered state to low temperatures below the critical temperature T_c and the kinetics of ordering are analyzed. We show that the approach to equilibrium is influenced by the effects of roughening fluctuation, domain-wall interactions, and pinning in two dimensions. These effects result in both quantitative as well as qualitative modifications of the usual theory of domain growth developed for three-dimensional systems. In two dimensions, domain growth rates become strongly temperature dependent, while domain-wall competition can result in a disordered structure characterized by the quenched-in domains of some average size. In this work simple domain geometries are studied using both an analytic domain-wall model as well as Monte Carlo simulations.

2 Simulation procedure

In this study the Monte Carlo method was utilized to simulate the grain growth. It was assumed that the lattice system is quenched from a high temperature disordered state (melting point) to a low temperature being below the critical temperature and then the growth phenomenon is initiated. In this method a computer image of the polycrystalline microstructure was created by mapping the continuum grain structure onto a discrete lattice.

The nearest-neighbor repulsions are accounted for by the mapping to the Potts model and the longer-range attractive interactions are transformed into spin-spin interactions with the Hamiltonian

$$H = \frac{J}{2}\sum_{i=1}^{N}\sum_{j=1}^{nn}\left(1-\delta_{S_i S_j}\right) \tag{1}$$

where S_i is one of the Q state of the spin on site i and δ is the Kronecker delta function. The sum is over nearest-neighbor spins on the Potts lattice and $J > 0$. We have studied reasonably large system sizes, up to 200x200 sites with periodic boundary conditions. Standard Monte Carlo methods were employed, using Glauber dynamic, with the transition probability W chosen to be

$$W = \begin{cases} \frac{1}{\tau} e^{-\Delta E/k_B T} & \Delta E > 0 \\ \frac{1}{\tau} & \Delta E \le 0 \end{cases} \tag{2}$$

where ΔE is the change energy caused by the change in spin orientation, K_B is the Boltzmann constant and T is the temperature. The constant τ sets the time scale for the Monte Carlo study. Since $1/\tau$ is like an attempt frequency, we expect it to have an Arrhenius temperature dependence for any particular experimental system, Thus $\tau \propto \exp(-Q'/k_B T)$ where Q' is an activation energy related to the coupling to the heat bath. In the following calculations, τ just sets the time scale. However, if $Q' \ll k_B T$, τ is independent of temperature for the growth kinetics. In any case, the temperature dependence introduced by τ, although important in a detailed comparison of theory and experiment, is unrelated to the cooperative interactions between the spins. Using Eq. (1), the transition probability W is computed and compared to a random number S ($0 \le S \le 1$) and the spin is changed if $W > S$; otherwise, the old configuration is retained. In the case of Kawasaki dynamics, the procedure is similar except that we randomly choose nearest-neighbour pairs of spins for exchange. The energy ΔE is computed before and after the spins are changed. The transition probability W' is defined by

$$W' = \frac{1}{\tau}\frac{e^{-\Delta E/k_B T}}{1+e^{-\Delta E/k_B T}}$$

As above, the exchange is carried out only if $W' > S$. The configurational averaging is obtained by averaging the data over many runs and we define the unit of time as one Monte Carlo step per spin (MCS) which corresponds to N microtrials or spin-flip attemps, where N is the total number of spins.

3 Circular domain kinetics

In contrast with the strip domain where the kinetics is dominated by fluctuations, an initially circular domain of one spin surrounded by an infinite sea of the other spin shrinks because of the deterministic force due to curvature. However, in contrast to the situation in three dimensions where fluctuations can be ignored, there is a strong temperature dependence to this shrinking rate. We thus write $R = R_0 + R_1 + R_2 + \ldots$, with R_0 (the deterministic part) independent of T, and R and R_2 being of order $T^{1/2}$ and T, respectively. In the following, the Boltzman constant is set to unity. Equating equal powers of T in the equations of motion we find

$$\dot{R}_0 = -\frac{J}{R_0} \tag{3}$$

$$\dot{R}_1 = \frac{JR_1}{R_0^2} + \frac{JR_{1\theta\theta}}{R_0^2} + \frac{\zeta}{R_0^2} \tag{4}$$

$$\dot{R}_2 = \frac{JR_2}{R_0^2} - \frac{JR_1^2}{R_0^3} + \frac{JR_{2\theta\theta}}{R_0^2} - \frac{2JR_1R_{1\theta\theta}}{R_0^3} - \frac{\zeta R_1}{2R_0^{3/2}} \tag{5}$$

The area of the minority domain is then computed as a function of time. Since

$$A = \frac{1}{2}\int \left\langle R^2(\theta)\right\rangle d\theta \tag{6}$$

only the angle-averaged values of R_0, R_1^2, and R_2 are needed. To solve these equations, we note that the solution of Eq. (1) is the deterministic solution

$$R_0^2(0) - R_0^2(t) = 2Jt \tag{7}$$

The equation for R_1 is linear and may thus be solved by Fourier transformation. We write

$$\left\langle R_1(0,t)R_1(\theta,t)\right\rangle = \sum_{n=-N}^{N-1} G_n e^{in\theta} \tag{8}$$

The cutoff N=πR is due to the discreteness of the lattice. Equation (2) then becomes

$$\dot{G}_n = \frac{2JG_n\left(1-n^2\right)}{R_0^2} + \frac{T}{4\pi R_0} \tag{9}$$

with the solution

$$G_n = \left[\left(\frac{R_0}{L_0}\right)^{2n^2-3} - 1\right]\frac{TR_0}{4\pi J\left(3-2n^2\right)} \tag{10}$$

The time rate of change of the area is

$$\dot{A} = \pi\frac{\partial}{\partial t}\left(R_0^2 + \sum_n G_n + 2R_0\overline{R}_2\right) \tag{11}$$

or

$$\dot{A} = 2\pi\left(-1 + \sum_n \frac{n^2 G_n}{R_0^2} + \frac{T}{8J}\right) \tag{12}$$

with

$$\overline{R}_2 = \frac{1}{2\pi}\int_0^{2\pi} R_2\, d\theta \tag{13}$$

We note that the time is still in units of the hopping time. Using the results for R_0, R_1 and R_2 we find (omitting terms of order 1/R)

$$\overline{\alpha} \equiv \frac{1}{\pi}\dot{A} \approx -2\left(1 - \frac{3T}{8J}\right) \tag{14}$$

4 Results

Numerical solutions of the equations of motion yield similar results for $\overline{\alpha}$, with some radius dependent corrections. The area thus decreases linearly in time, but with a temperature-dependent coefficient which decreases the shrinking rate as the temperature is decreased. We note that this effect is not due to critical fluctuations which are important only for $T \approx T_c$ but rather to roughening fluctuations of the domain boundary location.

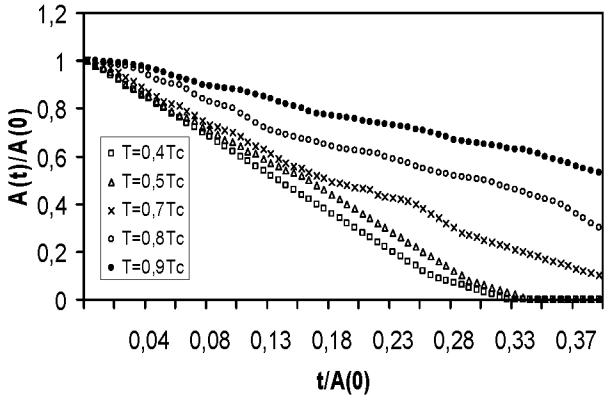

Figure 1 Normalized area $A(t)/A_0$ vs t/A_0 [A_0=A(0)] for circular domains of initial radius R=20 lattice constants at different temperatures obtained using Glauber dynamics. The temperature $T=0.9T_c$ (diamonds), $0.8T_c$ (circles), $0.7T_c$ (crosses), $0.5T_c$ (triangles), and $0.4T_c$ (squares). The data were averaged over 60 configurations for T/T_c =0.9 and 0.8, 40 configurations for T/T_c=0.7 and 0.5, and 20 configurations for T/T_c =0.4. The statistical error in $A(t)/A_0$ is comparable to the size of the symbols.

The simulations and the theoretical results can be compared quantitatively as well. Figures 1 and 2 show the linear time dependence of the area of the domain for several different temperatures. The approximate expression for the shrinking rate $\overline{\alpha}$ is plotted in Fig. 3 along with the results of Monte Carlo simulations for Glauber dynamics for an initial radius of 20 lattice spacings. The analytical results and the simulations have been fit at one point, since the overall time scale used in the simulation differs from that of the continuum theory.

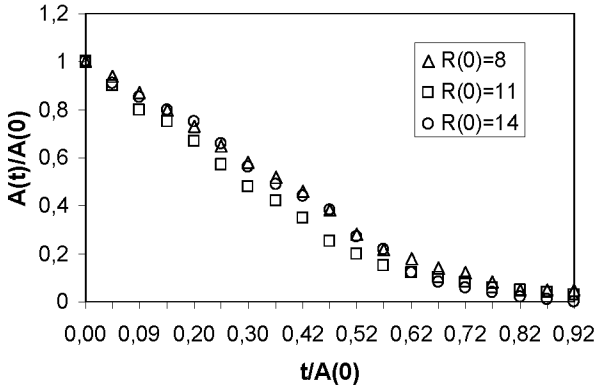

Figure 2 Normalized area $A(t)/A_0$ vs t/A_0 of initially circular domains using Kawasaki dynamics. The triangles, squares and circles correspond to circular domains of initial radii of R_0=8, 11 and 14 lattice constants [R_0=R(0)], respectively, at $T=0.6T_c$ averaged over 60 configurations.

In both cases, the hopping time τ has been set to unity; τ could provide some extrinsic, additional temperature dependence in real experimental situations. The agreement between theory and the simulation is excellent in the region T > J. In this region $\bar{\alpha}$ is linear in temperature due to the roughening effects. These fluctuations effectively increase the area at any given time, resulting in a decreased $\bar{\alpha}$ when compared with $\bar{\alpha}$ calculated from a deterministic theory. The deviation from the deterministic theory is not small, indicating the importance of these roughening effects.

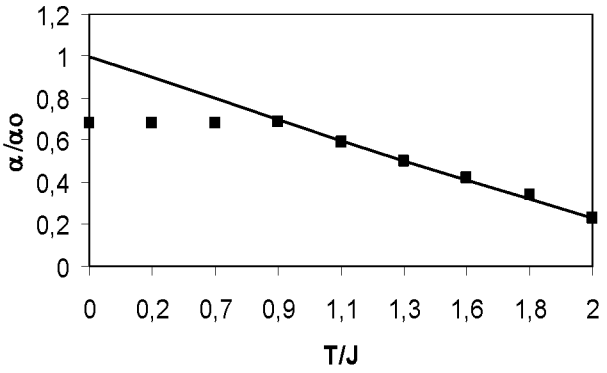

Figure 3 Shrinking rate $\alpha = \bar{\alpha}/\tau$ (see Eq. (14)) is plotted as a function of temperature normalized to the interaction energy J. The points are the results of Monte Carlo simulations using Glauber dynamics, and the line is the result of the roughening model described in the text. For both the simulations and the model calculation, the hopping time τ has been set to unity. The value for α obtained from the model calculation is normalized to its T=0 value.

At low temperature, the simulations show a temperature-independent value which is about 30% lower than that predicted by the extrapolation of the continuum theory to zero temperature. The reason for this discrepancy lies in the effects of the discreteness of both the space lattice and the spins, which do not allow a perfectly circular initial domain. Furthermore, the motion of the domain boundary must proceed in discrete steps in the simulations.

Thus when compared with a continuum theory for an initially circular domain, these discreteness effects result in some effective roughness, even at zero temperature. This effective roughness leads to the temperature-independent decrease of $\bar{\alpha}$ at low temperatures from the predicted theoretical values.

The simulations using Kawasaki dynamics yield a more complicated temperature dependence for $\bar{\alpha}$, since the effects of the exchange dynamics introduces another intrinsic temperature dependence for the shrinking rate α. This is shown in Fig. 4 where $\ln(\bar{\alpha})$ vs J/T is plotted. At the lowest temperatures, $\bar{\alpha}$ is temperature independent. For $0.3 \le T/T_c \le 0.6$, the Monte Carlo results fall on a straight line with a slope of 0.4. At higher temperatures, however, a strong deviation from linearity (which implies exponential temperature dependence for $\bar{\alpha}$) is found. In the inset of Fig. 4 $\alpha' \cong \bar{\alpha}\, e^{0.4\,J/T}$ is plotted versus T/T_c and a linear decrease of α' is found for $T > 0.6T_c$ as above. The deviation at $T = 0.95T_c$ from this linear behavior is presumably due to critical fluctuations; the domain walls lose their sharpness at $T \to T_c$. At fixed temperatures ($T = 0.8T_c$) we have also observed a weak but systematic dependence of α on the initial radius. As the initial radius is changed from 10 to 20 lattice constants, α increases by 15%. However, as the initial radius is further increased to 33 lattice constants, α decreases by 20%.

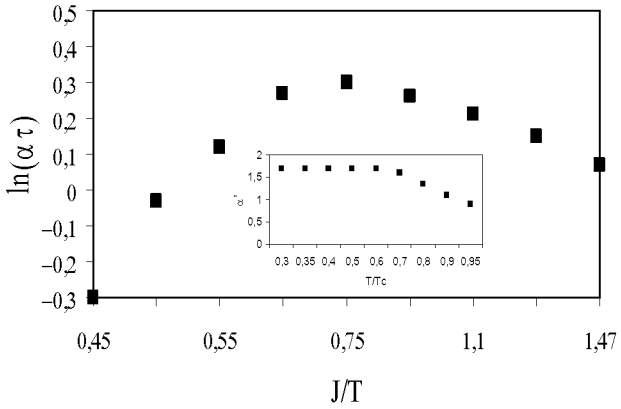

Figure 4 Logarithm of the slope α obtained from simulations using Kawasaki dynamics is plotted vs J/T. The inset displays $\alpha' = \alpha\tau e^{0.4J/kT}$ vs T/T_c.

Finally, we note that simulations of the kinetics of global quenches on two-component systems have resulted in temperature-dependent growth rates that are identical to those obtained here for the isolated domain studied here. This indicates the utility of our model domain studies where analytic models can be used to understand this temperature dependence in terms of roughening fluctuations of the domain walls. The fact that for the Ising model, the temperature dependence is the same for both the model study and the global quenches reflects the appropriateness of the simple geometry as being a typically nucleated domain.

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